

(19)



Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11) Publication number:

**0 446 911 A1**

(12)

**EUROPEAN PATENT APPLICATION**(21) Application number: **91103917.0**

(51) Int. Cl.<sup>5</sup>: **C07C 69/773**, C07C 255/55,  
C07C 331/26, C09K 19/20,  
C09K 19/30

(22) Date of filing: **13.03.91**

(30) Priority: **16.03.90 EP 90104964**  
**23.05.90 EP 90109781**

(43) Date of publication of application:  
**18.09.91 Bulletin 91/38**

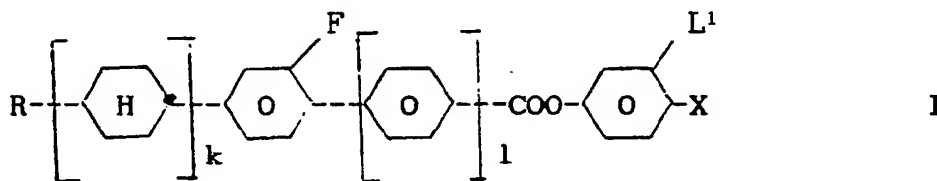
(84) Designated Contracting States:  
**DE GB**

(71) Applicant: **MERCK PATENT GESELLSCHAFT  
MIT BESCHRÄNKTER HAFTUNG**  
**Frankfurter Strasse 250**  
**W-6100 Darmstadt(DE)**

(72) Inventor: **Coates, David Dr.**  
**87 Sopwith Crescent Merley, Wimborne,**  
**Dorset BH21 3SW(GB)**  
Inventor: **Hittich, Reinhard Dr.**  
**Am Kirchberg 11**  
**W-6101 Modautal 1(DE)**  
Inventor: **Krause, Joachim Dr.**  
**Samuel-Morse-Strasse 14**  
**W-6110 Dieburg(DE)**  
Inventor: **Plach, Herbert Dr.**  
**Wingertsbergstrasse 5**  
**W-6100 Darmstadt(DE)**

(54) **Fluorobenzene derivatives.**

(57) The invention relates to fluorobenzene derivatives of the formula I



wherein

R denotes an alkyl residue of up to 12 C atoms wherein one or two non-adjacent CH<sub>2</sub> groups may also be replaced by -O- and/or -HC=CH-,

k is 0, 1 or 2,

l is 0 or 1,

L<sup>1</sup> is H or F, and

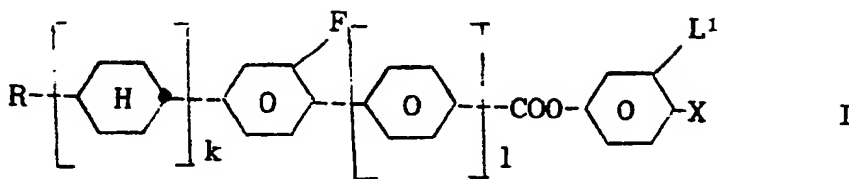
X denotes CN, NCS, F, Cl, CF<sub>3</sub>, OCF<sub>3</sub> or OCHF<sub>2</sub> with the provisos that

(a) L<sup>1</sup> denotes F if simultaneously k = 1, l = 0 and x = F or Cl and

(b) denotes 1 or k denotes 2 if simultaneously L<sup>1</sup> = F and X = CN.

**EP 0 446 911 A1**

The invention relates to fluorobenzene derivatives of the formula I



wherein

R denotes an alkyl residue of up to 12 C atoms wherein one or two non-adjacent CH<sub>2</sub> groups may also be replaced by -O- and/or -HC=CH-,

k is 0, 1 or 2,

l is 0 or 1,

L<sup>1</sup> is H or F, and

X denotes CN, NCS, F, Cl, CF<sub>3</sub>, OCF<sub>3</sub> or OCHF<sub>2</sub>, with the provisos that

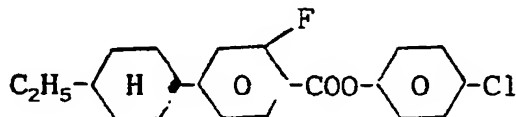
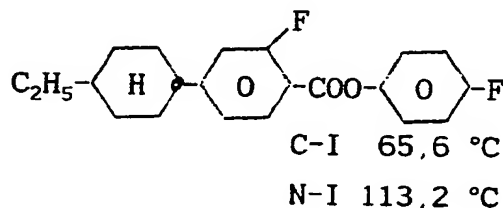
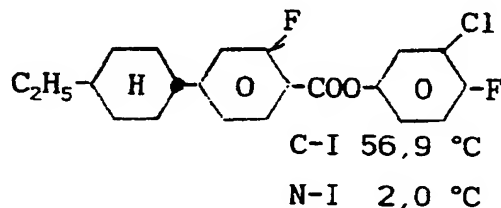
(a) L<sup>1</sup> denotes F if simultaneously k = 1, l = 0 and X = F or Cl and

(b) l denotes 1 or k denotes 2 if simultaneously L<sup>1</sup> = F and X = CN,

and also to liquid crystalline media being a mixture of at least 2 compounds, characterized in that at least one compound is a fluorinated benzene derivative according to formula I.

The invention was based on the object of discovering new stable liquid crystal or mesogenic compounds which are suitable as components of liquid crystalline media and, in particular, have advantageous values for optical and dielectric anisotropy combined with low viscosity and high nematogenicity.

Similar fluoro-chloro compounds are known from EP 0 219 975:

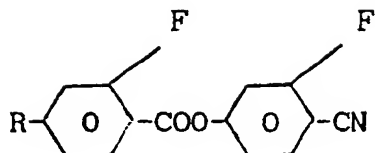


45

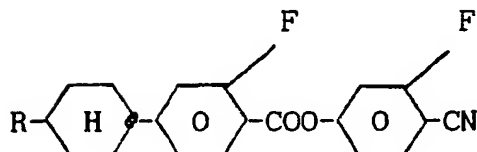
50

55

Similar fluoro-cyano compounds are known from G.W. Gray et al., MCLC, 172, 165 (1989):



and EP 0219975:



It has now been found that laterally fluorinated compounds of formula I are highly suitable as polar components of liquid crystalline media. In particular, they have especially advantageous values of optical and dielectric anisotropy and are not strongly smectogenic. It is also possible to obtain stable liquid crystal phases with a broad nematic mesophase range including a good deep temperature behaviour, low dependence of the threshold voltage on the temperature, a high resistivity and a comparatively low viscosity with the aid of these compounds.

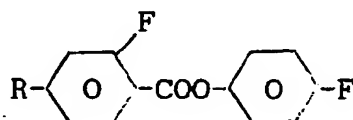
Depending on the choice of R, k, l, L<sup>1</sup> and X, the compounds of the formula I can be used as the base materials from which liquid crystal media are predominantly composed; however, it is also possible for compounds of the formula I to be added to liquid crystal base materials of other classes of compounds, for example in order to influence the dielectric and/or optical anisotropy and/or the viscosity and/or the nematic mesophase range of such a dielectric.

The compounds of the formula I are colourless in the pure state and are liquid crystalline in a temperature range which is favourably placed for electrooptical use. They are very stable towards chemicals, heat and light.

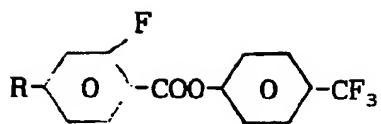
The invention thus relates to the benzene derivatives of the formula I, to liquid crystalline media with at least two liquid crystalline components, wherein at least one component is a compound of the formula I and to liquid crystal display devices containing such media.

Above and below, R, k, l, L<sup>1</sup> and X have the meaning given unless expressly indicated otherwise.

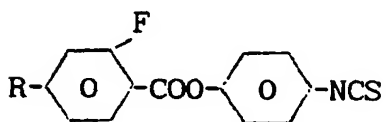
The compounds of the formula I include the preferred benzene derivatives of the formulae Ia to Im:



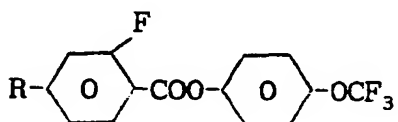
Ia



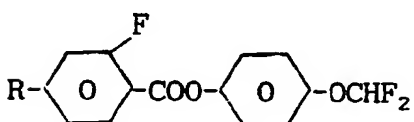
Ib



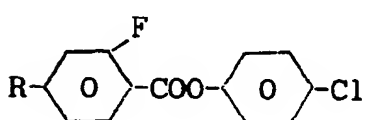
Ic



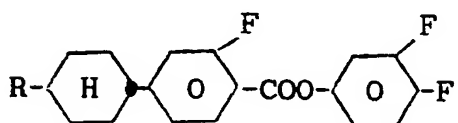
Id



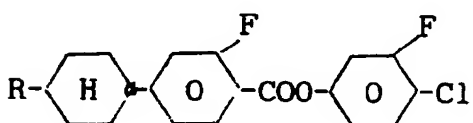
Ie



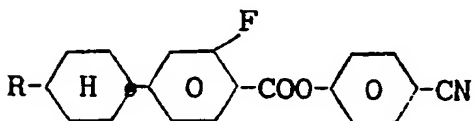
If



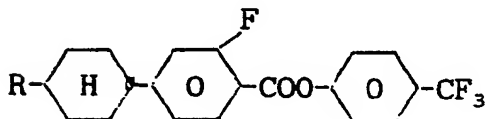
Ig



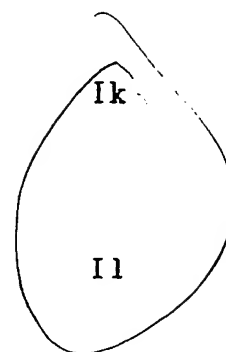
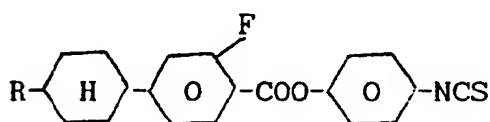
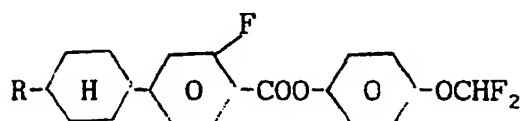
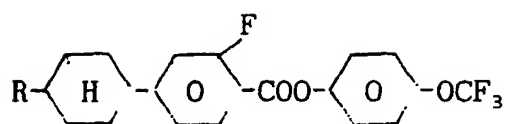
Ih



Ii



Ij



I m

Also preferred are the compounds of the formula I wherein  $k = 0, l = 1$  or  $k = 2, l = 0$  or  $k = 1, l = 1$ .

R is preferably alkyl, alkoxy, oxaalkyl or alkenyl and can exhibit a straight-chain or branched structure.

Alkyl or alkoxy preferably are straight-chain and have 2, 3, 4, 5, 6 or 7 C atoms. Accordingly they are preferably ethyl, propyl, butyl, pentyl, hexyl, heptyl, ethoxy, propoxy, butoxy, pentoxy, hexoxy or heptoxy, also methyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, methoxy, octoxy, nonoxy, decoxy, undecoxy, dodecoxy, tridecoxy or tetradecoxy.

Oxaalkyl is preferably straight-chain 2-oxapropyl (= methoxymethyl), 2-(= ethoxymethyl) or 3-oxybutyl (= 2-methoxyethyl), 2-, 3- or 4-oxapentyl, 2-, 3-, 4- or 5-oxahexyl, 2-, 3-, 4- 5- or 6-oxaheptyl, 2-, 3-, 4-, 5-, 6- or 7-oxaoctyl, 2-, 3-, 4-, 5-, 6-, 7- or 8-oxanonyl or 2-, 3-, 4-, 5-, 6-, 7-, 8-, or 9-oxadecyl.

Alkenyl is preferably straight-chain and has 2 or 10 C atoms. It is accordingly, in particular, vinyl, prop-1- or prop-2-enyl, but-1-, -2- or -3-enyl, pent-1-, -2-, -3- or -4-enyl, hex-1-, -2-, -3-, -4- or -5-enyl, hept-1-, -2-, -3-, -4-, -5- or -6-enyl, oct-1-, -2-, -3-, -4-, -5- -6- or -7-enyl, non-1-, -2-, -3-, -4-, -5-, -6-, -7- or -8-enyl or dec-1-, -2-, -3-, -4-, -5-, -6-, -7-, -8- or -9-enyl.

Compounds of the formula I containing a branched terminal group can occasionally be of importance because of an improved solubility in the customary liquid crystal base materials, but in particular as chiral doping substances if they are optically active.

Branched groups of this type as a rule contain not more than one chain branching. Preferred branched radicals are isopropyl, 2-butyl (= 1-methylpropyl), isobutyl (= 2-methylpropyl, 2-methylbutyl, isopentyl, (= 3-methylbutyl), 2-methylpentyl, 2-ethylhexyl, 2-propylpentyl, 2-octyl, isopropoxy, 2-methylpropoxy, 2-methylbutoxy, 3-methylbutoxy, 2-methylpentoxy, 3-methylpentoxy, 2-ethylhexoxy, 2-methylhexoxy, 1-methylhexoxy, 1-methylheptoxy (= 2-octyloxy), 2-oxa-3-methylbutyl, 3-oxa-4-methylpentyl, 4-methylhexyl, 2-nonyl, 2-decyl, 2-dodecyl, 6-methyloctoxy, oxy, 2-methyl-3-oxapentyl and 2-methyl-3-oxahexyl.

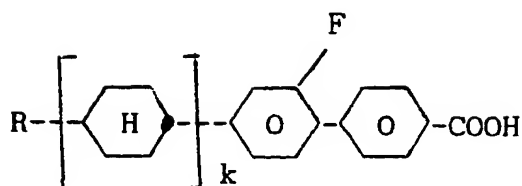
In the case of compounds with a branched terminal group R, formula I includes both the optical antipodes and racemates as well as mixtures thereof. k is preferably 1 or 2. Also preferred are compounds wherein  $k = 0$  and  $l = 1$ . L' is preferably F. X is preferably F, Cl,  $\text{CF}_3$ ,  $\text{OCF}_3$  or  $\text{OCHF}_2$ .

Of the compounds of the formula I and subformulae thereof, those in which at least one of the radicals contained therein has one of the preferred meanings given are preferred.

The compounds of the formula I are prepared by methods which are known per se, such as are described in the literature (for example in the standard works, such as Houben-Weyl, Methoden der Organischen Chemie Methods of Organic Chemistry, Georg Thieme Verlag, Stuttgart), and in particular under reaction conditions which are known and suitable for the reactions mentioned. Variants which are known per se and are not mentioned in more detail here can also be used in this connection.

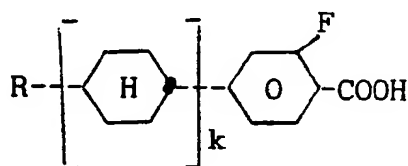
If desired, the starting materials can also be formed in situ, such that they are not isolated from the reaction mixture but are immediately reacted further to give the compounds of the formula I.

The carboxylic acids of the formula

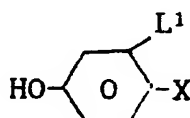


can easily be obtained from the known benzonitriles (German Offenlegungsschriften DE 31 36 624 and DE 30 42 391).

The carboxylic acids of the formula



and their reactive derivatives, e.g. the corresponding acyl chlorides, are well known in the art as well as the reaction conditions for esterification with phenols of formula

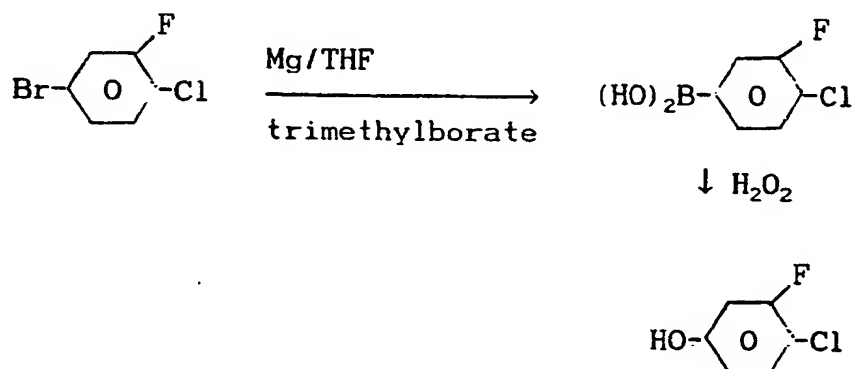


or their reactive derivatives.

Most of these phenols are known. All can be made by conventional methods.

A preferred route for preparation of the fluoro-chloro phenols is shown in the following scheme 1:

Scheme 1:



All starting materials are known or prepared in analogy to known starting materials.

55 Other routes are apparent to the skilled worker. All these steps and the corresponding reaction conditions are known to the skilled worker.

In addition to one or more compounds for formula I the liquid crystal media according to the invention preferably contain 2-40 components and in particular 4-30 components. Liquid crystal media being

composed of one or more compounds of formula I and 7-25 other components are especially preferred.

These additional components are preferably chosen from the nematic or nematogenic (monotropic or isotropic) substances; in particular from the classes of azoxybenzenes, benzylideneanilines, biphenyls, terphenyls, phenyl or cyclohexyl benzoates, phenyl or cyclohexyl cyclohexanecarboxylates, phenyl or cyclohexyl cyclohexylbenzoates, phenyl or cyclohexyl cyclohexylcyclohexanecarboxylates, cyclohexylphenylbenzoates, cyclohexylphenyl cyclohexanecarboxylates, cyclohexylphenyl cyclohexylcyclohexanecarboxylates, phenylcyclohexanes, cyclohexylbiphenyls, phenylcyclohexylcyclohexanes, cyclohexylcyclohexanes, cyclohexylcyclohexenes, cyclohexylcyclohexylcyclohexene, 1,4-bis-cyclohexylbenzenes, 4,4'-bis-cyclohexylbiphenyls, phenyl- or cyclohexylpyrimidines, phenyl- or cyclohexylpyridines, phenyl- or cyclohexyldioxanes, phenyl- or cyclohexyl-1,3-dithianes, 1,2-diphenylethanes, 1,2-dicyclohexylethanes, 1-phenyl-2-cyclohexylethanes, 1-cyclohexyl-2-(4-phenyl-cyclohexyl)-ethanes, 1-cyclohexyl-2-biphenylethanes, 1-phenyl-2-cyclohexyl-phenylethanes, optionally halogenated stilbenes, benzyl phenyl ethers, tolanes and substituted cinnamic acids.

The 1,4-phenylene groups of these compounds may be fluorinated.

The most important compounds which are possible constituents of liquid crystal media according to the invention can be characterized by the formulae 1, 2, 3, 4 and 5:

20	$R' - L - U - R''$	1
	$R' - L - COO - U - R''$	2
	$R' - L - OOC - U - R''$	3
	$R' - L - CH_2CH_2 - U - R''$	4
25	$R' - L - CC - U - R''$	5

In the formulae 1, 2, 3, 4 and 5 L and U may be equal or different from each other. L and U independently from each other denote a bivalent residue selected from the group consisting of -Phe-, -Cyc-, -Phe-Phe-, -Phe-Cyc-, -Cyc-Cyc-, -Pyr-, -Dio-, -G-Phe-, -G-Cyc- and their mirror images; in this compilation of residues Phe denotes unsubstituted or fluorinated 1,4-phenylene, Cyc trans-1,4-cyclohexylene or 1,4-cyclohexenylene, Pyr pyrimidine-2,5-diyl or pyridine-2,5-diyl, Dio 1,3-dioxane-2,5-diyl and G 2-(trans-1,4-cyclohexyl)-ethyl, pyrimidine-2,5-diyl, pyridine-2,5-diyl or 1,3-dioxane-2,5-diyl.

One of the residues L and U is preferably Cyc, Phe or Pyr. U preferably denotes Cyc, Phe or Phe-Cyc. The liquid crystal media according to the invention preferably contain one or more components selected from the compounds of formulae 1, 2, 3, 4 and 5 with L and U meaning Cyc, Phe and Pyr, said liquid crystal media further containing at the same time one or more components selected from the compounds of formulae 1, 2, 3, 4 and 5 with one of the residues L and U denoting Cyc, Phe and Pyr and the other residue being selected from the group consisting of -Phe-Phe-, -Phe-Cyc-, -Cyc-Cyc-, -G-Cyc-, said liquid crystal media containing in addition to this optionally one or more components selected from the compounds of formulae 1, 2, 3, 4 and 5 with L and U being selected from the group consisting of -Phe-Cyc-, -Cyc-Cyc-, -G-Phe- and -G-Cyc.

In a preferred subgroup of the compounds of formulae 1, 2, 3, 4 and 5 (subgroup 1) R' and R'' are independently from each other alkyl, alkenyl, alkoxy, alkenoxy with up to 8 carbon atoms. R' and R'' differ from one another in most of these compounds, one of the residues usually being alkyl or alkenyl. In another preferred subgroup of the compounds of formulae 1, 2, 3, 4 and 5 (subgroup 2) R'' denotes -CN, -CF<sub>3</sub>, -F, -Cl or -NCS while R' has the meaning indicated in subgroup 1 and is preferably alkyl or alkenyl. Other variants of the envisaged substituents in the compounds of formulae 1, 2, 3, 4 and 5 are also customary. Many such substances are commercially available. All these substances are obtainable by methods which are known from the literature or by analogous methods.

The liquid crystal media according to the invention preferably contain in addition to components selected from subgroup 1 also components of subgroup 2, the percentage of these components being as follows:

- subgroup 1: 20 to 90 %, in particular 30 to 90 %
- subgroup 2: 10 to 50 %, in particular 10 to 50 %

In these liquid crystal media the percentages of the compounds according to the invention and the compounds of subgroup 1 and 2 may add up to give 100 %.

The media according to the invention preferably contain 1 to 40 %, in particular 5 to 30 % of the compounds according to the invention. Media containing more than 40 %, in particular 45 to 90 % of the compounds according to the invention are further preferred. The media contain preferably 3, 4 or 5 compounds according to the invention.

5 The media according to the invention are prepared in a manner which is customary per se. As a rule, the components are dissolved in one another, advantageously at elevated temperature. The liquid crystal media according to the invention can be modified by suitable additives so that they can be used in all the types of liquid crystal display devices. Such additives are known to the expert and are described in detail in the literature (H. Kelker/R. Hatz, Handbook of Liquid Crystals, Verlag Chemie, Weinheim, 1980). For  
10 example, it is possible to add pleochroic dyestuffs to prepare colored guest-host systems or substances for modifying the dielectric anisotropy, the viscosity and/or the orientation of the nematic phases.

The following examples are to be construed as merely illustrative and not limitative. m.p. = melting point, c.p. = clearing point. In the foregoing and in the following all parts and percentages are by weight and the temperatures are set forth in degrees Celsius. "Customary work-up" means that water is added, the  
15 mixture is extracted with methylene chloride, the organic phase is separated off, dried and evaporated, and the product is purified by crystallization and/or chromatography.

Further are:

C: crystalline-solid state, S: smectic phase (the index denoting the typ of smectic phase), N: nematic phase, Ch: cholesteric phase, I: isotropic phase. The number being embraced by 2 of these symbols denotes the  
20 temperature of phase change.

#### Example 1:

##### Step 1

25 Magnesium (6.4 g, 0.263 moles) and THF (150 ml) were stirred under nitrogen. 1-bromo-3-fluoro-4-chlorobenzene (50 g, 0.239 moles) was added dropwise to the reaction mixture which was left to stir and reflux for 2 hrs. The reaction mixture was cooled to 0 °C, and trimethyl borate (30 ml, 0.263 moles) in THF (50 ml) was added keeping the temperature below 20 °C. The reaction mixture was left to stir in the ice  
30 bath for 30 mins, then quenched with acid solution (45 ml  $\text{CHCl}_3$ , 80 ml  $\text{H}_2\text{O}$ ). The reaction mixture was extracted into ether, washed well with  $\text{H}_2\text{O}$ , then brine and finally the ether was evaporated to afford the crude boronic acid. The boronic acid was dissolved in ether (165 ml) and 30 % hydrogen peroxide solution (30 ml, 0.263 moles) was added dropwise over 30 minutes. This was then left to stir at room temperature  
35 to remove excess  $\text{H}_2\text{O}_2$ . The reaction mixture was extracted with ether, washed well with  $\text{H}_2\text{O}$  then with ferrous sulphate to remove excess  $\text{H}_2\text{O}_2$ . The organic layer was extracted with 1 M NaOH solution and the aqueous phase acidified with  $\text{CH}_3\text{SO}_3\text{H}$ . The acidic solution was re-extracted into ether, dried and run down. The crude product was distilled to afford the phenol, bp 64 °/2.5 mbar.

##### Step 2

40 The phenol (1.84 g, 0.013 moles) and p-(trans-4-n-propylcyclohexyl)-o-fluoro-benzoic acid (0.0143 moles) were dissolved in DCM (30 ml) with trifluoroacetic anhydride (2 ml, 0.0143 moles). This was left to stand for 16 hrs. The reaction mixture was added to  $\text{H}_2\text{O}$  (30 ml), the organic layer was washed with  $\text{NaHCO}_3$  solution, then with  $\text{H}_2\text{O}$  and dried and run down. After customary work-up 3-fluoro-4-chlorophenyl  
45 p-(trans-4-n-propylcyclohexyl)-o-fluoro-benzoate is obtained.

#### Examples 2 to 13:

In place of the used carboxylic acid and the fluoro-chlorophenol other starting materials can be used for  
50 preparation of the following compounds in analogy to example 1:



	R	k	l	L <sup>1</sup>	X
5	(2) n-pentyl	1	0	F	Cl
	(3) n-propyl	1	0	F	F
	(4) n-pentyl	1	0	F	F
	(5) n-pentyl	0	0	H	CN
10	(6) n-pentyl	0	0	H	CF <sub>3</sub>
	(7) n-pentyl	0	0	H	NCS
	(8) n-pentyl	0	0	H	OCF <sub>3</sub>
15	(9) n-pentyl	0	0	H	OCHF <sub>2</sub>
	(10) n-pentyl	1	0	H	CF <sub>3</sub>
20	(11) n-pentyl	1	0	H	OCF <sub>3</sub>

25

30

35

40

45

50

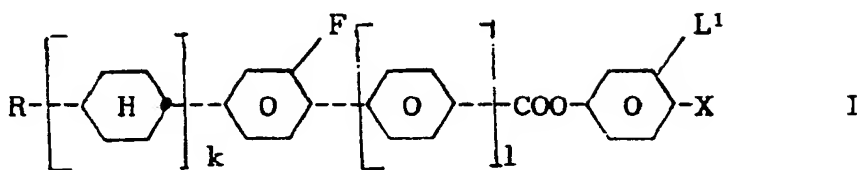
55

	(12)	n-pentyl	1	0	H	OCHF <sub>2</sub>	
5	(13)	n-pentyl	1	0	H	NCS	
	(14)	n-propyl	1	0	H	CF <sub>3</sub>	
	(15)	n-propyl	1	0	H	OCF <sub>3</sub>	
10	(16)	n-propyl	1	0	H	OCHF <sub>2</sub>	
	(17)	n-propyl	1	0	H	NCS	
	(18)	n-propyl	1	0	H	CN	
15	(19)	n-propyl	2	0	H	F	
	(20)	n-propyl	2	0	H	Cl	
	(21)	n-propyl	2	0	H	CF <sub>3</sub>	
20	(22)	n-propyl	2	0	F	F	
	(23)	n-propyl	2	0	F	Cl	
	(24)	n-propyl	0	0	H	Cl	
25	(25)	n-pentyl	0	0	H	Cl	
	(26)	n-propyl	0	1	H	CN	
30	(27)	n-pentyl	0	1	H	CN	C 118.9 N 195.4 I
	(28)	n-propyl	0	1	H	Cl	
	(29)	n-pentyl	0	1	H	Cl	
35	(30)	n-propyl	0	1	H	OCHF <sub>2</sub>	
	(31)	n-pentyl	0	1	H	OCHF <sub>2</sub>	
	(32)	n-propyl	0	1	F	Cl	
40	(33)	n-pentyl	0	1	F	Cl	
	(34)	n-propyl	0	1	H	OCF <sub>3</sub>	
	(35)	n-pentyl	0	1	H	OCF <sub>3</sub>	
45	(36)	n-propyl	1	1	F	F	
	(37)	n-pentyl	1	1	F	F	
50	(38)	n-propyl	1	1	H	OCF <sub>3</sub>	

(39)	n-pentyl	1	1	H	OCF <sub>3</sub>
(40)	n-propyl	1	1	H	CN
(41)	n-propyl	1	1	F	Cl

# 10 Claims

## 1. Fluorobenzene derivatives of the formula I



wherein

- R denotes an alkyl residue of up to 12 C atoms wherein one or two non-adjacent CH<sub>2</sub> groups may also be replaced by -O- and/or -HC=CH-,
- 25 k is 0, 1 or 2,
- l is 0 or 1,
- L¹ is H or F, and
- X denotes CN, NCS, F, Cl, CF<sub>3</sub>, OCF<sub>3</sub> or OCHF<sub>2</sub>, with the provisos that
- (a) L¹ denotes F if simultaneously k = 1, l = 0 and X = F or Cl and
- 30 (b) l denotes 1 or k denotes 2 if simultaneously L¹ = F and X = CN.

2. Liquid crystalline medium being a mixture of at least two compounds, characterized in that at least one compound is a benzene derivative according to claim 1.

35 3. Liquid crystal display device, characterized in that it contains a liquid crystalline medium according to claim 2.

4. Electrooptical display device, characterized in that it contains a liquid crystalline medium according to claim 2.

40

45

50

55



European  
Patent Office

## EUROPEAN SEARCH REPORT

Application Number

EP 91 10 3917

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
D,X	MOLECULAR CRYSTALS AND LIQUID CRYSTALS. vol. 172, 1989, MONTREUX CH pages 165 - 189; G.W. Gray et al.: "The synthesis and transition temperatures of some fluoro-substituted 4-cyanophenyl and 4-cyanobiphenyl-4'-yl pentyl- and 4-butoxybenzoates" * page 170, table 1 * - - - -	1	C 07 C 69/773 C 07 C 255/55 C 07 C 331/26 C 09 K 19/20 C 09 K 19/30
D,X	EP-A-0 219 975 (CHISSO CORP.) * claims 2-4, 8 * - - - -	1-3	
X	PATENT ABSTRACTS OF JAPAN vol. 8, no. 279 (C-257)(1716) 20 December 1984, & JP-A-59 148752 (CHISSO CORP.) 25 August 1984, * the whole document * - - - -	1	
X	EP-A-0 354 434 (MERCK PATENT) * claim 1 * - - - -	1	
X	WO-A-8 703 870 (MERCK PATENT) * claims 1, 5 * - - - - -	1,4	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C 07 C C 09 K
The present search report has been drawn up for all claims			
Place of search Berlin		Date of completion of search 26 June 91	Examiner PROBERT C.L.
<div>CATEGORY OF CITED DOCUMENTS</div> <div>X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention</div> <div>E: earlier patent document, but published on, or after the filing date D: document cited in the application L: document cited for other reasons</div> <div>&amp;: member of the same patent family, corresponding document</div>			